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Tunable Excimer Lasers

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Abstract

The wide bandwidth nature of the rare-gas halide excimer transitions allow reasonable tuning of the laser oscillation wavelength that makes it useful for a number of applications. At the same time this wide bandwidth makes narrow band operation difficult and special techniques are needed to insure narrow frequency lasing as well as absolute frequency resettability. I will discuss briefly some of the classical frequency narrowing techniques and then go on to some recent work that require lasers of special frequency characteristics for special applications including KrF laser fusion.

Introduction

I will first briefly discuss the limits of tunability and of desired output frequency spectrum characteristics due to the physical limitations that are characteristic of the particular excimer laser transition. Specific techniques for frequency narrowing are similar to those of flash-lamp pumped dye lasers. The difficulties of the shorter operating wavelengths are primarily related to the availability of comparable dispersive elements as compared to those for the visible spectrum. However, this situation is getting better everyday. The many techniques for frequency narrowing and tuning as well as the benefits of long-pulse laser operation on frequency narrowing have previously been discussed [1]. I will give some recent results on frequency narrowing for short pulse lasers to give a feel for what our expectations can be for excimer laser devices. Finally, I would like to discuss some recent work on two frequency operation that have applications to laser radar detection of atmospheric species [2] and to the shaping of broad-band coherent sources [3] in the kilojoule regime for KrF laser fusion experiments.

Physical Limitations

The ability of a particular rare-gas halide transition to tune depends in large part on the nature of its ground state potential. Although the ground state (X) is basically dissociative, van der Waal's forces can create a potential well in varying degrees in the different rare-gas halide molecules. These vary from the cases of KrF [4], ArF [5], KrCl [6] and XeBr [7] which are dissociative with relatively non-structured, wide bandwidth fluorescence to slightly bound ground states for XeCl (approx. 200 cm^{-1} well) [6] with fluorescence to five or more lower vibrational states and broad-band lasing to four of these states, and finally to tightly bound states in XeF which has a fairly deep well of 1065 cm^{-1} giving structured but still fairly broad fluorescence [8] although resulting in discrete narrow line lasing.

As an example of a smooth fluorescence spectrum the full width at half maximum bandwidth for KrF is 400 cm^{-1} (Fig.1). The fluorescence linewidth is not completely determined by the shape of the ground state potential; there are significant (although not large) contributions due to higher vibrational levels of the upper laser state. Because of the higher vibrational state contributions and the shape of the ground state potential the fluorescence line shape is not completely symmetrical, but the B-X molecular transition by and large behaves as a homogeneously broadened system.

This, however, is not true as one tries to tune through the total fluorescence bandwidth of the XeCl B-X system. The transitions to each vibrational lower state behaves homogeneously but radiation wavelengths to different ground state vibrational transitions behave inhomogeneously. This observation implies that the lower state vibrational levels do not mix well in gain times under consideration (25-35 ns). The problem does not lie in the ability to tune through the fluorescence bandwidth in an oscillator but in the subsequent amplification to high energies. This is because the gain at one frequency does not decrease homogeneously when the medium is saturated by fields at another frequency when the two frequencies access different ground state vibrational levels. In single pass amplification this shows up as an inability to suppress amplified spontaneous emission at other wavelengths other than the injected wavelength. When the amplification is performed in a regenerative amplifier configuration the problem can become much more severe. It becomes necessary to pick a large enough magnification for the unstable resonator optics so that the unattenuated gain at other wavelengths than the injected wavelength can not go into laser oscillation.

Although it is not possible to go into great detail here, it is necessary to say a few words regarding the apparent contradiction of the above statements to recent interpretations of some femtoseconds gain dynamics measurements performed by S. Szatmari and F.P. Shafer [9,10]. Szatmari and Shafer used 220 fs pulses to saturate the XeCl and KrF B-X transitions and then followed with a small signal variable delayed gain probe. They observe in XeCl a sharp drop off in gain due to the saturating field with a very quick recovery of 65% of the gain within 53 ps. Full recovery of the gain takes a few nanoseconds due to repumping of the upper state by the discharge. The initial gain recovery can be interpreted as refilling of the upper state via rotational relaxation or/and collisional dissociation of the lower state. For a 53 ps lower state dissociation rate one will require a dissociation rate constants in collisions with the buffer gas (density at 10^{19} cm^{-3}) at 20 times gas kinetic (10^{-10} cm^3/s). In lieu of similar initial gain recovery times in KrF which does not have a bound lower state, it is likely that the effect is due to repopulation of the upper state from rotational relaxation. I feel the lower state dissociation rate may be substantially longer. Szatmari and Shafer measure quantum beats between the two lower states with a period of 1.28 ps and a damping time constant of 12 ps. They believe this damping time constant to represent the population redistribution time of the lower states. In light of the fact that transitions to different lower states behave inhomogeneously, the lower state population redistribution time need to be slow compared to the dissociation time so that the lower state populations are not mixed. This is really not inconsistent with the femtosecond studies as the damping time constant of the quantum beats only require dephasing collisions and not collisions that change states.

A second limitation is the presence of discrete line absorption within the gain bandwidth of the broad-band transition. The existence of an absorber in KrF which initially we thought due to CF_2 is shown not to be so from Fig.1. Presently, the speculation is that this is due to excited state absorption which means it will be extremely difficult to minimize and impossible to eliminate. This absorption presents problems in tuning and broad-band lasing. ArF happens to be in the region of the Shuman-Runga band of O_2. Large frequency holes develop in the lasing line if there is air within the optical cavity or the propagating path [4].

Frequency Control of Excimer Lasers

The fact that excimer laser transitions are an extremely efficient and powerful source of ultraviolet radiation makes it an ultraviolet source of choice for applications that require moderate to high energies or average powers. The very high single photon energies make it a unique source for photochemical applications as the single photon energy is sufficient to break many chemical bonds. The wide bandwidth nature of the transitions typically allows for tunability of over 30 angstroms as well as wide bandwidth oscillation that eliminates spacial coherence structures which are problems in certain applications.

The requirement for narrow linewidth and frequency resettability is not only limited to hitting absorption resonances in molecules and atoms but for very important industrial processes such as semiconductor photolithography. The stringent frequency requirements for photolithography results from the frequency dependent nature of the index of refraction of optical elements. Thus, when line widths of the size of the wavelengths of the illuminating source is to be written on semiconductor devices the source linewidths and the absolute laser wavelength cannot drift more than 1 to 2 picometers. For those who like to think in other units as this author does, this translates to .06 to .12 cm^{-1} or 1.8 to 3.6 GHz linewidth.

Frequency narrowing and tuning have been done with etalons, prisms and gratings. Usually etalons are angle rather than pressure tuned. For the wide band transitions of the excimer lasers generally more than one etalon are needed to narrow the linewidth sufficiently for many applications. Our experience has shown that when three etalons are used to achieve a particular linewidth, the insertion loss becomes so high that unless the etalons really are of very high quality competition between the line narrowed feedback signal and amplified spontaneous emission becomes a severe problem. Tuning is generally very complicated and require synchronization of all the angles of the etalons. This is generally done with computer control. Prisms are used both as dispersion elements and as beam expanders. Often beam expanders are used in conjunction with large area gratings to insure that the total length of the grating is utilized. The use of gratings are typically in two configurations coupled with other frequency selective elements within the cavity. Some of these configurations have been discussed previously by Sze et al. [1]. One configuration is to use the grating in glancing incidence. The first order scatter from the grating is then fed back into the cavity for frequency narrowing and control. Tuning is accomplished by tuning the feed-back mirror. When the grating is very large (the larger the area of a grating the narrower the linewidth) the glancing incidence angle is very shallow. In these situations the first order scatter can be too weak to effectively compete against amplified spontaneous emission. The solution to this is to use beam expanders so that the grating can be completely flooded by the laser at more reasonable gracing incidence angles to insure greater light scattering off the first order of the grating. The limit of this is to expand the beam large enough so that the grating can be completely flooded with the grating angle set at Littrow configuration where the first order scatter is directly fed back to the laser.

Figure 2 gives an example of the linewidth after grazing incidence tuning at KrF (248 nm) wavelength by a two inch long grating having 3290 grooves per millimeter and sending it subsequently through a single pass amplifier with resultant output power of 25 millijoules. The linewidth achieved here is .2 cm^{-1}. Using another grating in the Littrow configuration for the first order feed back mirror of the glancing incidence grating, we achieve .14 cm^{-1} which nearly meets the requirements for a photolithography source. The rings are the laser output after passing through a 1 cm^{-1} analyzing etalon. Frequency resettability or the need of the narrowed line source not to drift by more than 1 picometer is presently done by locking the cavity to a wavemeter set to a specific frequency.

It is possible to tune the excimer lasers to more than one wavelength with an etalon. This is done by setting two etalon modes equally spaced about line center if the gain is symmetrical; otherwise, one straddles line center so that the two etalon modes access two regions of equal gain. As one tunes off this equal gain situation, the higher gain etalon mode will take over and only one wavelength region will oscillate. In practice it is very easy to keep the laser operating in two modes. Figure 3 shows the oscillating wavelength for KrF as an intra-cavity etalon of 225 cm^{-1} separation and 50% reflectivity is angle tuned through the total bandwidth of the etalon. There is another important effect which is illustrated in the figure, and that is the effect to tuning due to the existence of a small absorption some 85 cm^{-1} off line center as was shown in Fig.1. We see that the absorption prevents lasing in that region. This puts important limits to the operation of wide bandwidth lasing in KrF. Possible applications for this two wavelength source is its use as an input into large amplifier systems to produce broad bandwidth laser for fusion research and as a possible source for differential LIDAR applications [10]. Figure 4 shows two frequency operation using a 118 cm^{-1} etalon of 80 % reflectivity. (a) shows the output out of the oscillator and (b) shows the output after single pass amplification with energies per pulse at 25 millijoules. Higher etalon reflectivities will result in narrower linewidths. We can tune one wavelength to an absorption of the species under investigation while the other wavelength will measure other atmospheric scattering (Mies or Rutherford). Tuning needs to be done by changing the etalon separation. When the two frequency operation is very closely spaced more than one etalon will be required. Figure 5 shows two wavelength operation of 2 cm^{-1} (60 GHz) separation by using 75 cm^{-1} and 2 cm^{-1} intra-cavity etalons. (a) shows the ring patterns using a 10 cm^{-1} analyzing etalon and (b) shows the temporal waveform after propagation through the amplifier.

The idea for generating broad-band sources after amplification through the Aurora fusion amplifier begins with a frequency shaped pulse out of the oscillator. The oscillator pulse is generated using a fairly low reflectivity intra-cavity etalon tuned for two frequency operation. The etalon separation will be the desired width of the broad-band source. The use of a low reflectivity etalon is to establish enough radiation between the etalon wavelengths so that amplification at the higher gain regions near line center and amplification at the lower gain regions at the etalon wavelengths will reach the same intensity after passage through the amplifier chain. A propagation code [11] and experimental verification of the code [3] have been developed and studied. The results of the code calculations for the propagation of different bandwidth sources through the Aurora laser system, have been undertaken [12] and shows the 100 cm^{-1} bandwidths are easily obtainable. 200 cm^{-1} bandwidths are theoretically obtainable with additional filtering etalons in the early stages of amplification. However, in reality the bandwidths are limited to below 170 cm^{-1} due to the presence of the absorber mentioned earlier.

Discussion

We have shown that most of the excimer laser transitions are tunable over a range as large as 30 angstroms. Because of the wide gain bandwidth of the transitions normal operations give nearly 20 cm^{-1} (1.25 angstroms) linewidths. The need to narrow this linewidth for certain applications call for the insertion of dispersion elements into the cavity of the oscillator. The insertion loss usually means small energies out of the oscillator and generally there is a need for subsequent amplification of the line narrowed source. WE saw that certain excimer transitions behave inhomogeneously and one has to be careful that the magnification of regenerative amplifier optics must be kept high enough to keep other wavelengths from oscillating. WE see that the manipulation of the frequency is not limited to merely line-narrowing and tuning but can include two wavelength operation and ultra-broad-band lasing as well as other frequency characteristics for specific applications.

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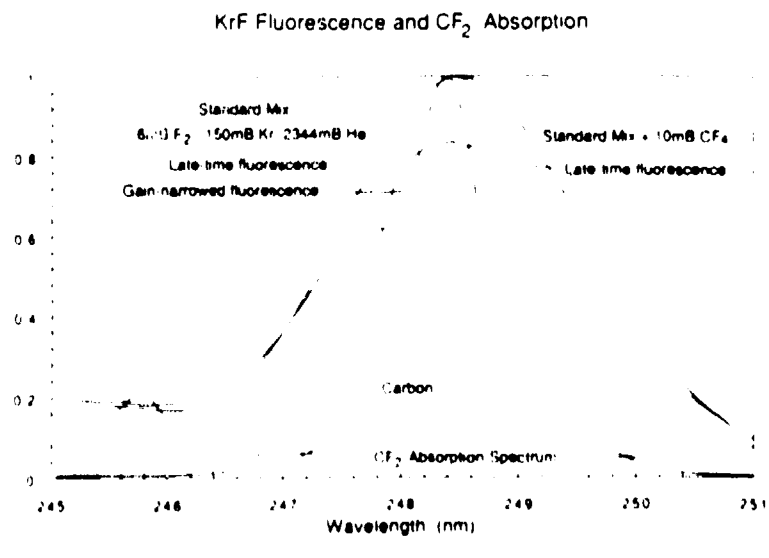


Figure 1. KrF fluorescence and CF_2 absorption spectra during discharge and in late time (approx. 1 microsecond). Curtesy of Andy McGowen of the Los Alamos National Laboratory.

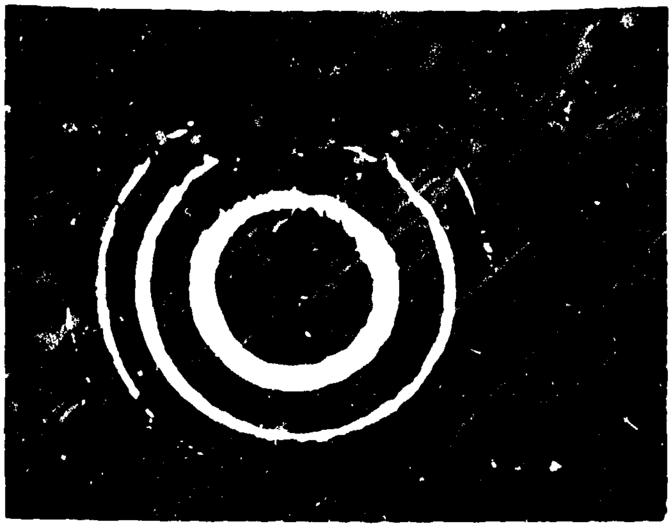


Figure 2. Grazing incidence grating line narrowing in KrF with a 3290 g/mm holographic grating. Observed with a 1 cm^{-1} etalon.

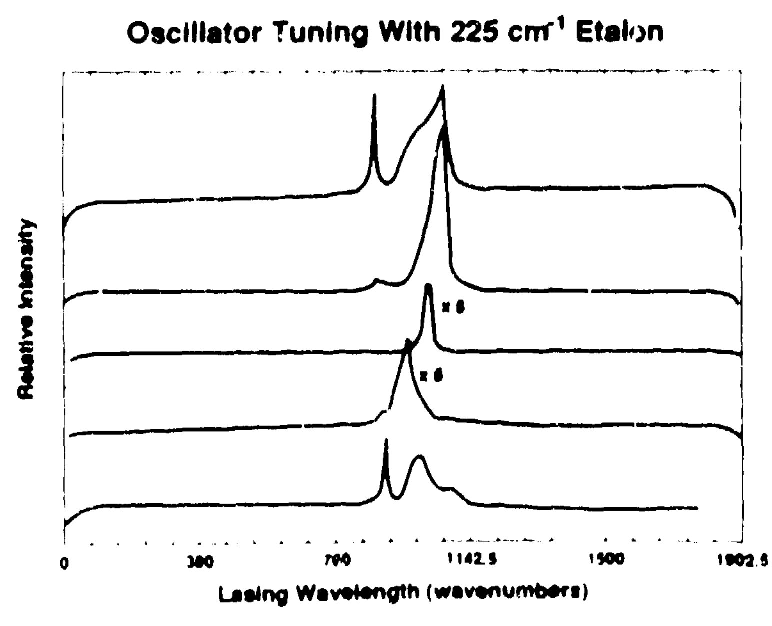
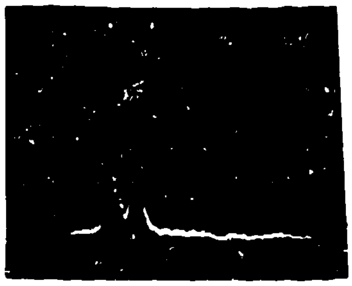


Figure 3. Wavelength tuning from two to single laser modes with a 225 cm^{-1} etalon of 50 % reflectivity.

(a)



(b)

Figure 4. Two frequency operation in KrF with a 118 cm^{-1} etalon of 80 % reflectivity. (a) spectral output from oscillator. (b) spectral output after a single pass through an amplifier giving 25 mj per pulse energy.

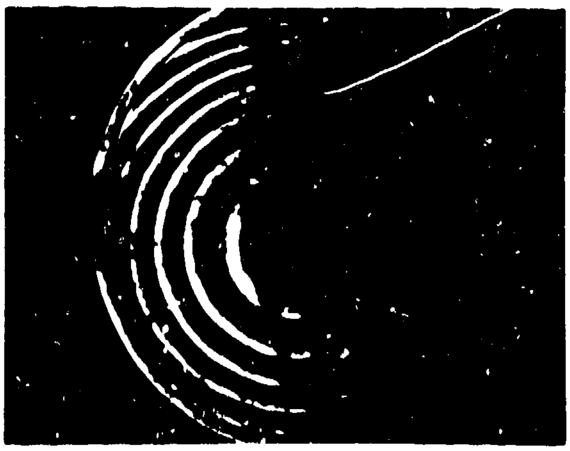


Figure 5. Two wavelength operation in KrF with 2 cm^{-1} (60 GHz) separation using two angle tuned intracavity etalons of 75 cm^{-1} and 2 cm^{-1} separation.